SINGLE-FREQUENCY CW OPTICAL PARAMETRIC OSCILLATORS: DEVICES AND APPLICATIONS

S. SCHILLER, J. SCHOSER, C. BRAXMAIER, K. BENCHEIKH,
U. STRÖßNER, A. PETERS, J. MLYNEK
*Optik-Zentrum Konstanz and Fakultät für Physik M696, Universität Konstanz,
D-78457 Konstanz, Germany, http://quantum-optics.physik.uni-konstanz.de*

P. DE NATALE
Istituto Nazionale di Ottica and LENS, Largo E. Fermi, 6 I-50125 Firenze, Italy

We describe the state-of-the-art in single-frequency cw OPOs, with emphasis on systems with high long-term stability. Doubly-resonant and singly-resonant OPOs are discussed and their performance summarized. First spectroscopic applications are presented and future perspectives given.

Introduction

Although the potential of cw OPOs has been recognized for more than three decades, only recently have the devices matured sufficiently to become useful for practical applications. Progress has been spurred by improvements and new developments in several areas during the last years: novel periodically poled (PP) crystals, powerful single-frequency pump sources, durable and broad-band dielectric coatings, frequency stabilization techniques, and novel device configurations and optimization.[1]

The motivation for pushing the development of single-frequency cw OPOs is that their properties can be superior to those of certain presently available tunable sources, such as lead-salt diode lasers, color center lasers, molecular lasers (CO overtone laser), and difference frequency generators. The properties to be considered are: emission range, output power, power stability, linewidth, absolute frequency stability, continuous frequency tunability, ease of tuning to particular frequencies, reliability, user-friendliness.

Fig.1 shows recent progress with respect to the emission range and the ability to oscillate without mode-hops. With the introduction of multi-grating PP crystals in 1995 the spectral coverage has been greatly extended, and now covers essentially the whole range from 0.7 to 4 μm. For spectroscopic applications, a crucial requirement is absence of mode-hops; the figure shows that this has been achieved with both doubly- and singly-resonant OPOs.

This review presents OPO systems that exhibit stable single-frequency behavior and gives examples for the above specifications. Doubly-resonant OPOs are discussed first. In the second part, singly-resonant OPOs are discussed both in general and then with focus on a particular type.

Proc. XIV Int. Conf. Laser Spectrosc. (World Scientific, 1999),p.217-26

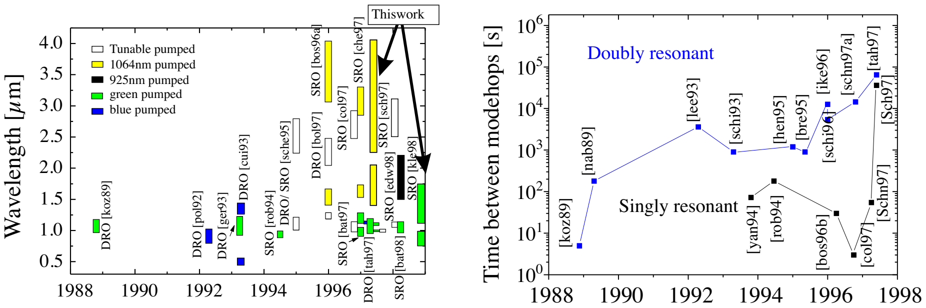

Figure 1. Cw OPO performance evolution as reported in the literature. Left: emission ranges of widely tunable OPOs (no distinction between single-frequency and non-single-frequency devices). Right: Maximum time between mode-hops observed for single-frequency OPOs.

Doubly-resonant OPOs

Historically, doubly-resonant OPOs (DROs) were the first to be developed, because of the lower threshold requirements. The characteristic feature of the DRO is that it is very sensitive to changes in the external parameters that influence the oscillation frequencies, the pump frequency ν_p, the crystal temperature T and the OPO cavity length contribution in air, L_a. The maximum permissible excursions in these parameters for keeping oscillation on a given signal/idler pair favorable compared to oscillation on any other pair can be calculated as a function of the nonlinear material, output wavelengths and cavity length. For a type-I interaction, typical tolerances are $\delta L_a \sim 0.5\,\text{nm}$, $\delta T \sim$ several mK, and $\delta\nu_p \sim$ few MHz.

The problem of these low tolerances has been essentially eliminated by active stabilization of the OPO cavity length.[2] An error signal is obtained from the resonating waves that carries information about their detuning from cavity modes. The cavity length is adjusted for optimum resonance of the oscillating signal/idler pair. In this way even large changes of crystal temperature and of pump frequency can be compensated (provided they are sufficiently slow). The tolerances increase to typically $\delta\nu_p \sim 100\,\text{GHz}$ and $\delta T \sim 0.2\,\text{K}$. Excursions in these parameters do, however, cause changes in the signal/idler frequencies, which may be undesirable or purposely applied to tune the output frequencies. The tolerances can be regarded as maximum tuning ranges for one-parameter tuning. For pump tuning, $\partial\omega_{s,i}/\partial\omega_p \simeq \omega_{s,i}/\omega_p$, while temperature tuning is weak.[3]

Experimentally, we have achieved mode-hop free oscillation over many hours with two similar devices.[3] Fig.2 shows the frequency stability of a MgO:LiNbO$_3$-DRO pumped by a monolithic Nd:YAG laser resonantly dou-

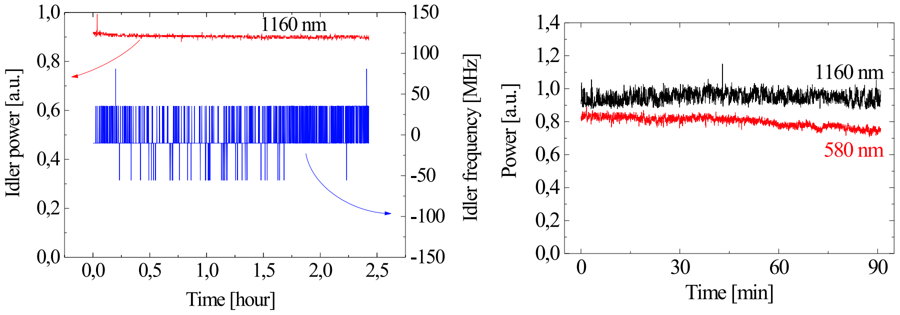

Figure 2. Left: Frequency stability of a 532 nm-pumped type-I DRO. Right: Power stability of idler wave at output of a fiber and of the resonantly doubled wave.

bled in another MgO:LiNbO$_3$-external cavity. This pump source has very low frequency drift and a linewidth below 10 kHz. The OPO frequency drift is below the resolution of the wavemeter employed (± 30 MHz), consistent with the low pump laser drift and the tuning coefficients.

Tuning of the output frequencies of the cavity-length-stabilized DRO is best done with a frequency-tunable pump laser. So far, pump lasers with maximum continuous tuning range of 19 GHz were used, with up to 9.4 GHz continuous tuning of the OPO output.[3] Much higher mode-hop-free tuning ranges should be possible, however. Gibson et al.[4] circumvented the limited tuning range of their Nd:YLF laser pumping a KTP-DRO. They covered 220 GHz by combining tuning over 4 GHz via the pump frequency with 9 GHz wide mode-hops induced externally and with angle tuning.

The linewidth of signal and idler waves is very narrow if a narrow-linewidth pump is used. Initially the linewidth of DROs was studied operating very close to degeneracy, so that the beat between signal and idler could be measured.[5] More recently, an upper limit for the linewidth of a DRO has been found by scanning the DRO frequency across a mode of a high-finesse cavity. The FWHM of the line was 40 kHz, limited by the cavity finesse.[3]

The outstanding spectral features of the DRO make it worth while to frequency-convert the radiation to the visible if an application requires it. We have resonantly doubled the idler wave of the above DRO in an additional cavity containing MgO:LiNbO$_3$, obtaining an output power of a few 100 μW at 580 nm for future use in high-resolution atomic spectroscopy. Fig.2 (right) shows that the whole system of three cascaded nonlinear resonators (in two different rooms connected via optical fiber) can be operated with acceptable power stability.

Finally, it should be mentioned that DROs have been demonstrated to be very efficient, with up to 81% of pump power converted to signal and idler waves,[6] and to generate output powers of up to 400 mW.[7] Both results were achieved in frequency-stable operation.

Singly-resonant OPOs

The original motivation to develop cw singly-resonant OPOs was to take advantage of their lower sensitivity to perturbations of the external parameters as compared to an unstabilized DRO. Since the threshold of a SRO is about two orders of magnitude higher than that of a DRO, the first demonstration of a cw SRO occurred only relatively recently.[8] Our own motivation for studying them was to pursue a device type that could furnish a wide emission range without optics change.

Since the frequency stability properties of SRO are of interest, it is useful to present a simple model for the sensitivity of the SRO to external perturbations, based on a discussion of the parametric gain. Consider a cavity in which one of the parametrically generated waves (frequency ω_1, mode number N_1) is resonated. Let L_a be the round-trip propagation distance in air, and L_c that within the crystal. We assume that the allowable parameter tolerances are given by the condition that the gain of the currently resonated mode remains larger than the gain of the next longitudinal mode N_1+1. For larger deviation of the parameters, oscillation on this neighboring mode would be favored. Since the gain is approx. proportional to $\mathrm{sinc}^2(\Delta k L_c/2)$, gain equality implies that the wavevector mismatch satisfies

$$\Delta k(\omega_p+\delta\omega_p, T, L_a, N_1) = -\Delta k(\omega_p+\delta\omega_p, T, L_a, N_1+1) \qquad (1)$$

for the maximum one-sided deviation (tolerance) of pump frequency, $\delta\omega_p$ and similarly for δL_a, δT. The mismatch is $c\Delta k = n_p(\omega_p,T)\omega_p - n_1(\omega_1,T)\omega_1 - n_2(\omega_2,T)\omega_2$, where $\omega_2=\omega_p-\omega_1$ and ω_1 (and possibly ω_p) is a function of T, L_a, N_1 via the resonance condition $\omega_1(L_a+n_1(\omega_1,T)L_c(T)) = 2\pi c N_1$. Considering that ω_p, T, L_a may not all be independent parameters, by Taylor-expansion we obtain the conditions

$$\begin{aligned}
&[\tilde n_p-\tilde n_2+(\tilde n_2-\tilde n_1)\partial\omega_1/\partial\omega_p+\Gamma\partial T/\partial\omega_p]\,\delta\omega_p = (\tilde n_2-\tilde n_1)\Omega/2\\
&[(\tilde n_p-\tilde n_2)\partial\omega_p/\partial L_a+(\tilde n_2-\tilde n_1)\partial\omega_1/\partial L_a]\,\delta L_a = (\tilde n_2-\tilde n_1)\Omega/2\\
&[(\tilde n_p-\tilde n_2)\partial\omega_p/\partial T+(\tilde n_2-\tilde n_1)\partial\omega_1/\partial T+\Gamma]\,\delta T = (\tilde n_2-\tilde n_1)\Omega/2\ , \qquad (2)
\end{aligned}$$

where Ω is the free spectral range, $\Gamma=\omega_p\,dn_p/dT-\omega_1\,dn_1/dT-\omega_2\,dn_2/dT$, $\tilde n_x = n_x(\omega_x)+\omega_x dn_x/d\omega_x$. These equations apply to all SRO types. To specialize them to a particular type, only those conditions are considered that refer to an independent parameter, and the partial derivatives, where they are nonzero, are calculated using the corresponding resonance condition for ω_1 and possibly for ω_p.

For example, in the simple SRO, with non-resonant pump wave, ω_p, L_a, T are all independent parameters, and the nonzero derivatives are $\partial\omega_1/\partial L_a = -\omega_1/L(\omega_1)$, $\partial\omega_1/\partial T = -\omega_1 L'(\omega_1)/L(\omega_1)$ with the effective length $L(\omega_x) = L_a + L_c\tilde{n}_x$, and $L'(\omega_x) = d(n_x(\omega_x, T)L_c(T))/dT$ is the temperature derivative of the crystal optical path length for the wave x. Typical values are $\delta L_a \simeq$ half a resonant wavelength, $\delta\nu_p \sim$ several 100 MHz, and $\delta T \sim$ tens of mK. While these are orders of magnitude better than for an unstabilized DRO, the cavity length will typically drift sufficiently strongly for mode-hops to occur on time scales of minutes.[8] To prevent this, several approaches are possible. Intracavity etalons may be used,[9,10] which is equivalent to enforcing a larger effective value for Ω in Eqs.(2).

Alternatively, the cavity length may be actively stabilized. This approach can be usefully combined with a strong reduction in threshold in the pump-resonant SRO (PR-SRO).[11] The idea is to use a frequency-stable pump and to resonate it in the SRO cavity by controlling its length L_a and thus significantly reducing its drift.[12] Using the above argument, we can determine the tolerances for a PR-SRO. Since the length L_a is controlled by the locking servo, the independent parameters are now the pump frequency ω_p and the crystal temperature T. The second condition in Eq.(2) is irrelevant and the non-zero derivatives are

$$\partial\omega_1/\partial\omega_p = \omega_1 L(\omega_p)/\omega_p L(\omega_s)\ ,\quad \partial\omega_1/\partial T = \omega_1(L'(\omega_p) - L'(\omega_1))/L(\omega_1)\ . \quad (3)$$

For example, for a 1064 nm-pumped PR-SRO using PP-lithium niobate (PPLN) with 3.3 μm idler $\delta\nu_p = 2.5$ GHz, $\delta T = 35$ mK, while for a 532 nm-pumped PP-lithium tantalate (PPLT) OPO with 1.48 μm idler $\delta\nu_p = 1$ GHz, $\delta T = 7$ mK. These tolerances are much lower than for a length-stabilized DRO, However, they can be satisfied even over long times by frequency-stable pump lasers and accurate crystal temperature control. The tolerances increase the further away from degeneracy the PR-SRO is operated.

By locking the cavity length to the pump frequency, the stability of the pump frequency is transferred to the cavity optical path length of the cavity and therefore also to the resonated frequency ω_1. By photon energy conservation, the idler frequency will also be stable. The pump frequency and temperature being independent parameters, their drift or deliberate tuning leads to signal/idler tuning, with coefficients given by Eqs.(3). The tuning ranges in this model are equal to twice the tolerances derived above.

We now turn to a brief description of the results obtained with PR-SRO devices built in our group. As pumps we use again monolithic Nd:YAG lasers,

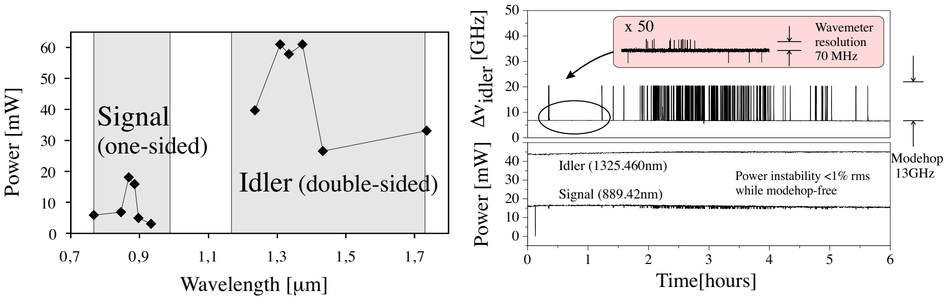

Figure 3. Left: Output power on signal and idler branch of the 532 nm-pumped PR-SRO. Right: Long-term measurement of idler frequency and output powers. A slow drift in pump frequency or crystal temperature causes the signal frequency to hop between two cavity frequencies spaced by 4 free spectral ranges, with corresponding jumps of the idler.

at 1064 nm and at 532 nm. The nonlinear crystals are PPLN and PPLT multi-gratings, respectively. The OPO cavities are configured in a semi-monolithic fashion, with one cavity mirror formed by the appropriately coated flat end face of the crystals. The temperature of the crystals is actively stabilized. Oscillation thresholds were as low as 135 mW for the 1064 nm-pumped device and 76 mW for the 532 nm-pumped one. This shows that with sufficient effort invested in low-loss coatings, thresholds comparable to those of DROs are reachable. For the 532 nm-pumped PPLT OPO, the threshold is influenced by the substantial crystal loss at the pump wavelength. In addition, power dependent thermal shifts of the cavity resonance occur, and careful adaptation of the cavity lock servo is required.

For the 1064 nm-pumped PPLN device, we achieved tuning over 1.45-1.99 μm and 2.29-4.0 μm. Output powers as high as 230 mW for the non-resonant idler were obtained, limited only by available pump power, with a photon conversion efficiency of 68%.[13] The dependence of output power on pump power was found to be in good agreement with theory.[11]

Fig.3 (left) shows in grey the emission range of the 532 nm-pumped PR-SRO. We note that the output powers vary substantially across the emission range due to variation of pump coupling, conversion nonlinearity and of the cavity loss due to the coatings and/or the crystal. At present, we have reached mode-hop free operation for almost one hour with this device (Fig.3, right).

Two 1064 nm-pumped PR-SROs have been routinely operated without mode-hops for over 10 h. Fig.4 (left) shows that the device tolerates substantial pump frequency drifts, as expected; the signal frequency tracks the pump frequency with the expected tuning coefficient. It can also be inferred that the frequency fluctuations of the signal *not* due to the pump frequency fluc-

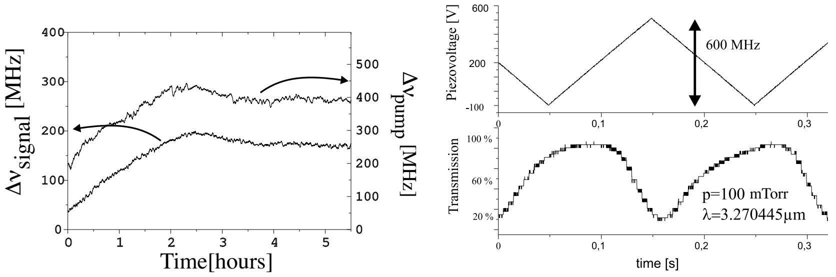

Figure 4. Left: Long-term measurement of 1064 nm pump and 1.57 μm signal frequencies with two high-resolution wavemeters. Signal frequency drift is almost entirely due to pump frequency drift. Right: Top: pump frequency control ramp; bottom: partial idler absorption line by methane; idler frequency range is about 200 MHz.

tuations are at a level below a few tens of MHz.

We have noticed for both these and the 532 nm-pumped device that when mode-hops do occur, the frequency change is often not a single but several free spectral ranges, indicating that spurious etalons are present in the standing-wave cavities. This effect improves stability, but hinders easy tuning of the PR-SRO to arbitrary emission frequencies. This is a process that requires careful adjustment of crystal temperature and pump frequency and can take several hours. However, it is possible to turn off the pump laser overnight and obtain the same oscillation frequency upon turning it on the next day.

Relatively rapid tuning of the PR-SRO output is possible if a fast cavity lock is used. Fig.4 (right) shows that a few 100 MHz can be covered in 50 ms, by using fast pump laser tuning. With slow tuning, a maximum range of 2 GHz has been reached. So far, only an upper limit for the linewidth of a PR-SRO could be given, 160 kHz.[14] A value similar to that of the pump laser, 10 kHz, is expected for both signal and idler.

We also built a breadboard-mounted portable PR-SRO and operated it on different continents.

Applications

The most important application for single-frequency OPOs is spectroscopy. Of wide interest is the generation of radiation in the mid-IR range (3 - 4 μm), where vibrational spectroscopy of the fundamental (and hence strongest) mode of the hydrogen bond in molecules can be performed. The potential for applications is large, ranging from frequency metrology to basic molecular physics and physical chemistry to more applied studies in the fields of environmental monitoring (trace gas analysis), earth sciences (isotope spectrometry), plant, animal and human biology (physiological process monitoring via molecular gas emissions).

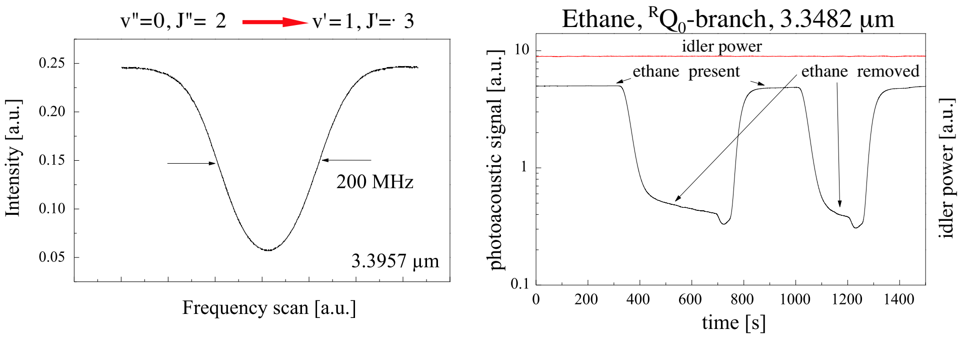

Figure 5. Spectroscopy with a PR-SRO. Left: Absorption line of HCl at 3.3957 μm. Recording time was approx. 1 min. Right: Detection of ethane injected into a photoacoustic cell using photoacoustic spectroscopy.

First demonstrations of spectroscopy with single-frequency OPOs were reported in early 1998 by a few groups.[3,4,9,13,15] As an example, Fig.5 (left) shows a transition in low-pressure HCl gas obtained with a 1064 nm-pumped PR-SRO, where the Doppler-broadened regime was reached.[13] The line exhibits good signal-to-noise ratio, due to the low intensity noise of the OPO.

An application for OPOs which makes use of several of their features is trace gas detection for applications in biology and medicine. Here the concentration of specific molecular compounds in air is to be monitored continuously over time scales between minutes and days. This can be accomplished using photoacoustic spectroscopy. The requirements for the laser are: sufficiently high power (tens of mW), reliable oscillation (i.e. mode-hop-free) with frequency stability better than the linewidth of the pressure-broadened molecular transition, and, for high sensitivity, the ability to tune to the strongest molecular transition lines.

We have used a 1064 nm-pumped PR-SRO to demonstrate that single-frequency OPOs show significant potential to be used for photoacoustic trace gas monitoring.[15] Fig.5 (right) shows a result of a test experiment in which known concentrations of ethane in nitrogen carrier gas were injected into a photoacoustic cell. Mode-hop-free oscillation of the OPO allowed continuous monitoring of the ethane concentration, and the low OPO intensity noise permitted to reach a detection sensitivity (0.5 ppb) comparable to that achieved with a conventional source, a CO-overtone laser, but at a much lower power level (40 mW vs. 1 W). Significant potential for improvement is expected by increasing OPO output power.

For applications in ultra-high resolution spectroscopy, the already low linewidth and frequency instability of OPOs must be further reduced. The best method is to stabilize the OPO output frequency to a stable reference. To demonstrate the feasibility of this method, we have stabilized the idler of a DRO to an ultra-stable cryogenic reference cavity, using the frequency of the pump laser as frequency control. The absolute frequency instability reached was estimated to be below 1 kHz.[3]

Summary and outlook

At this stage in OPO development, highly favorable specifications have been reached: a huge emission range with a single pump laser, high output power, conversion efficiencies of tens of percent, good short- and long-term power stability, linewidths below 40 kHz by using appropriate pump lasers, mode-hop-free operation with signal/idler frequency drifts below 50 MHz over an hour, and fast tunability over several hundred MHz.

The main conclusion from the above work is that both the DRO and the PR-SRO concept lead to output radiation with linewidth and frequency stability comparable to that of the pump laser. The choice of source type depends on the application. For a very wide spectral coverage, notably in the mid-IR, a PR-SRO with resonant signal is more suitable since the necessary coating bandwidth is reduced. However, the spectral coverage of DROs can be extended significantly with special coatings.[16] The PR-SRO is in principle more suited also if high output power is desired on either signal or idler, since it is fully available for use if the conjugate wave is resonated.

If a pump laser of lower power is available or desirable, a low threshold must be achieved. An analysis[11] shows that this is more easily minimized with a DRO, by making the mirrors HR for both signal and idler at the expense of conversion efficiency. For a large continuous frequency tuning range on both signal and idler (tens of GHz), a DRO appears more suitable. If radiation near half the pump frequency is desired, this will imply a DRO, unless a type-II nonlinear crystal with polarization-splitting elements in the cavity is used.

We note that the development of other OPO types, e.g. SROs containing intracavity etalons, is also progressing significantly.[17]

Future device development has many avenues to follow. DROs with wide emission range based on the higher nonlinearity PP materials should be pursued, especially for emission around 3 μm for molecular spectroscopy. Wide continuous frequency tuning of a DRO with a tunable diode laser is a distinct possibility. Pumping DROs and PR-SROs with a pump laser that is frequency-stabilized to a reference is expected (see above) to yield a substantial improvement in signal/idler frequency stability. Furthermore, precise measurements of OPO linewidths are desirable. An important practical aspect that deserves attention is the development of automatic tuning systems for OPOs, which will reliably and quickly generate desired frequencies and make OPOs more easily usable. Further directions are to extend the emission range by frequency conversion of OPO output to shorter wavelengths[18] and raising output powers. Suitable commercial single-frequency pump sources with multi-Watt power levels are available. For example, we characterized a 5 W frequency-doubled Nd:YVO_4 laser (Coherent "Verdi") by beat frequency measurements with a frequency-doubled Nd:YAG laser. A short-term (1 ms) linewidth of 100 kHz, a frequency jitter of several MHz and a frequency drift of about 100 MHz per hour were found. Since the laser is not frequency-tunable, appropriate OPO types must be used if frequency-tunable output is desired.

Near-term goals in applications of OPOs are saturation spectroscopy and two-photon spectroscopy of molecular vibrational transitions for a variety of molecular physics studies. A search for new IR molecular optical standards can be undertaken. For trace gas detection efforts should be directed towards improving frequency agility and demonstrating field use of portable cw OPOs.

In conclusion, we can expect many new uses to be made of single-frequency OPOs in the near future, both in spectroscopy and for general spectral measurements.

We acknowledge the important participation of K. Schneider, P. Kramper, R. Al-Tahtamouni, A. Beirer, O. Pradl, J.-P. Meyn, R. Wallenstein, F. Kühnemann, A. Hecker, A. Martis and W. Urban. This work was supported by BMBF grant 13N7025/8, the Optik-Zentrum Konstanz, the Hess Program of the German Science Foundation and the Lion Foundation.

References

1. See Appl. Phys. B **66**, Vol.6 (1998) and previous spec. issues.
2. A.J. Henderson et al., Opt. Lett. **20**, 1029 (1995).
3. R. Al-Tahtamouni et al., Appl. Phys. B **66**, 733 (1998).
4. G.M. Gibson et al., Opt. Lett. **23**, 40 (1998).
5. D. Lee and N.C. Wong, J.O.S.A. B **10**, 1659 (1993).
6. G. Breitenbach et al., J.O.S.A.B **12**, 2095 (1995).
7. M. Bode et al. , Opt. Comm. **148**, 117 (1998).
8. S.T. Yang et al., Opt. Lett. **18**, 971 (1993).
9. P. E. Powers et al., Opt. Lett. **23**, 159 (1998).
10. 1064 nm-pumped PPLN SRO, Coherent Inc., California (1997).
11. For the theory, see S. Schiller et al., to appear in J.O.S.A.B (1999).
12. K. Schneider et al., Opt. Lett. **22**, 1293 (1997).
13. K. Schneider et al., OSA TOPS Vol.19, p. 256 (OSA, 1998).
14. K. Schneider and S. Schiller, Appl. Phys. B. **65**, 775 (1997).
15. F. Kühnemann et al., Appl. Phys. B **66**, 741 (1998).

16. M. Tsunekane et al., Appl. Opt. **37**, 6459 (1988).
17. K. Boller et al. CLEO 1999.
18. W.R. Bosenberg et al., Opt. Lett. **23**, 207 (1998).